Combustion Research Facility News

Second Generation Distributed-Feedback Dye Laser Yields NO-Fluorescence Decay Rates

Time-resolved spectroscopy of combustion species often requires lasers that are continuously tunable, stable, and have subnanosecond time resolution and moderate spectral resolution (e.g. <10 GHz). Building on an earlier design, Paul Schrader and Roger Farrow have developed a second-generation distributed-feedback dye laser (DFDL) that better meets these requirements. Their earlier instrument was used to acquire laser-induced fluorescence (LIF) and degenerate four-wave mixing spectra and to study rotational energy-transfer rates of hydroxyl radicals in various bath gases (see CRF News, Vol. 20, No. 6). They have used the new instrument to measure NO-fluorescence decay rates in diffusion flames and in so doing found that current computational models underpredict NO quenching rates.

The improved geometry (Figure 1) of the new DFDL allows symmetrical pumping, which improves tuning and beam-pointing stability. As in the earlier model, the split beams in the newer version are reflected by tilt mirrors into the dye cell where they cross. The lasing wavelengths are determined by the spacing of the grating, and the DFDL wavelength is tuned by adjusting the angles of the tilt mirrors to vary the crossing angle of the pump beams. A 110-ps pulsewidth Nd:YAG laser pumped the DFDL at 532 nm and at a repetition rate of 20 Hz.

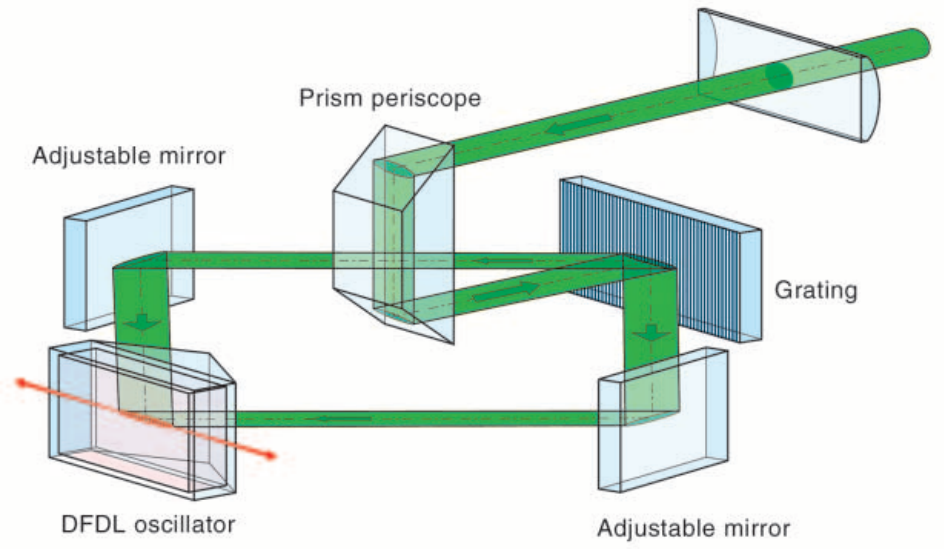

Figure 1. The distributed-feedback dye laser (DFDL) has a prism periscope that allows a symmetrical pumping geometry, which improves tuning and beam-pointing stability.

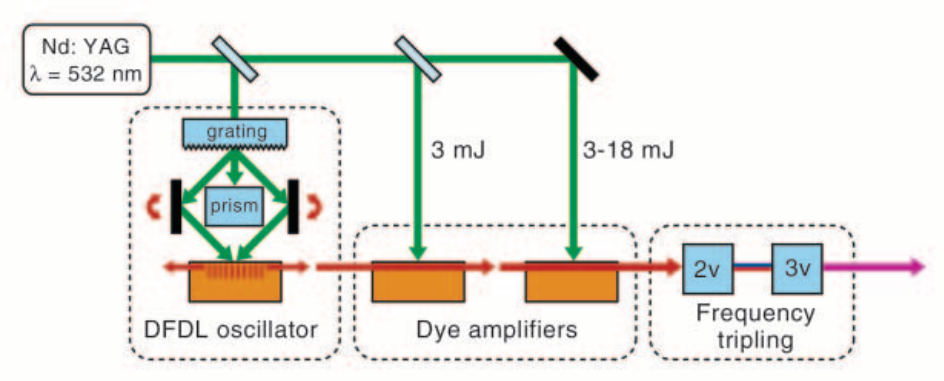

Figure 2. The new frequency-tripled DFDL-based laser is a significantly improved version of the first DFDL. Pulses are shorter and can be varied from 20 to 50 ps. Pulse jitter is much lower and wavelength tuning is smoother and faster owing to a new nanometer-resolution stepping motor. The simplified optical system allows high gain, single-pass amplification. After frequency tripling the wavelength is 226 or 230 nm, the pulse time is 55 ps, and the energy is about 400 μJ.

The DFDL output pulse was amplified in two dye amplifiers pumped by 532-nm pulses derived from the same Nd:YAG laser (see Figure 2). The use of a short pump pulse eliminated the need for a dye-cell quencher and complicated bow-tie amplifiers as in the previous DFDL, which used a 2-ns pump pulse. The DFDL amplified pulse energy was 1 mJ. The pulse duration was 34 ps, and the linewidth was a nearly transform limited 20 GHz.

Much of our understanding of the way pollutants are formed in combustion has come from the marriage of advanced laser diagnostics to predictive models. One of the remaining gaps in our understanding derives from our inability to predict the formation and removal of NO, particularly in fuel-rich and nonpremixed combustion. In direct injection engines, which offer important technological advantages, there is not enough time for complete mixing prior to ignition. Thus, knowledge of the formation and fate of NO under nonpremixed conditions has immediate practical importance.

Laser-induced fluorescence techniques can be used to obtain NO concentration profiles in atmospheric-pressure flames as a means of testing kinetic schemes used in combustion models. However, knowledge of the effective NO fluorescence lifetime is necessary to determine absolute NO concentrations. Under typical conditions the NO lifetime ranges from 1–4 ns, and can be directly measured using DFDL excitation and a microchannel plate photomultiplier tube. Working with Prof. Volker Sick and graduate student Josh Driscoll from the University of Michigan, the Sandia researchers have measured fluorescence in methane/air diffusion flames to obtain fluorescence decay rates (see Figure 3).

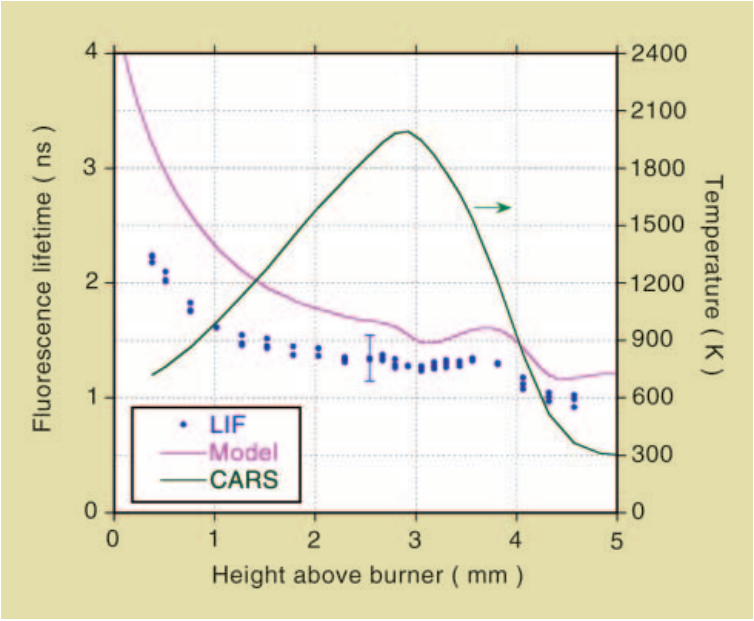

Figure 3. A comparison of measured and predicted fluorescence lifetimes of excited NO show that while the shapes of the plots are similar, the measured lifetimes are from 25-40% shorter than predicted. The CARS plot indicates the flame temperature at various locations in the flame.

Preliminary findings are that current quenching models may overpredict the NO fluorescence lifetimes by as much as 40%. Future work will include study of NO-doped flames, oxygen-atom LIF, and CH LIF profiles.

People

Norwegian visitors together with Sandians outside the Combustion Research Facility. From left to right are Lars Sørum (SINTEF Energy), Jarle Halmoy (Norwegian University of Science and Technology (NUST)), Chris Shaddix (Sandia), Inge Saanum, Christian Somme, Trym Ekrem, Oyvind Hunseid (all from NUST), Prof. Franz Winter (Sandia visitor from Vienna Technical University), and Linda Blevins (Sandia).

Professor Jeffrey A. Gray (center) of Ohio Northern University recently concluded a sabbatical visit to the CRF. While at the CRF, Prof. Gray collaborated with Tom Settersten (right) and Roger Farrow (left) on the development of infrared-UV four-wave-mixing methods for the detection of combustion intermediates.

This summer the CRF hosted 15 summer interns from a variety of educational institutions from all parts of the country to work on projects that ranged from web building to studies of the decomposition of energetic materials. Pictured from left to right are Brian Hart of Hamline University, St. Paul MN; David Goldsmith, Granada High School, Livermore; Trey Cauley, Iowa State University, Ames IA; Chris Herdman, Cornell Co-op, Ithaca, NY; Sepp Hammer, UC Berkeley; Chris Calderon, Purdue University, West Lafayette, IN; Alec Barlow, Amador Valley High School, Pleasanton; Kylie Smith, Notre Dame University, South Bend, IN; Kelsey Nyholm, Livermore High School, Janine Scott, Livermore High School; Melinda Cromie, UC Davis; Danielle Wiese-Smith, UC Berkeley; Grace Goelzer, UC Davis; David Rengel, Cornell University, Ithaca, NY, and Albert Castello, University of the Pacific, Stockton, CA.

CRF News Notes

The CRF and the International Truck and Engine Co. Collaborate to Measure EGR Distribution

Bob Green of the Engine Combustion group and Laura Ricart-Ugaz from the Advanced Combustion and Controls Department at International Truck and Engine Company recently collaborated to measure the exhaust gas recirculation (EGR) distribution in the intake system of a V-8 Diesel engine that is currently under development at International. Bob, along with his infrared-absorption, EGR diagnostic (see CRF News v. 21, n. 6, Nov/Dec 1999), traveled to Melrose Park, IL to set up for experimental measurements in an engine development test cell.

During a two-week effort, Bob and Laura interfaced the diagnostic and data acquisition system with the engine so that proper temporal phasing was established; acquired steady-state data for a number of operating modes that are representative of prescribed test cycles; and finally measured the EGR distribution during load transients imposed at a constant speed to simulate a rapid acceleration or deceleration. A quick look at the results indicates that very good data have been obtained; comprehensive data reduction is proceeding.

Laura worked at the CRF in 1997 while a graduate student at the University of Wisconsin. She spent six months working with John Dec on adapting the University of Wisconsin version of the KIVA II code to match experimental results from the Sandia/Cummins single-cylinder research diesel engine.

Fuel Cell Delivers Power to Mine Vehicle Locomotive

Jay Keller reports that initial tests on a fuel-cell-powered mine vehicle locomotive have demonstrated power generation and transfer to the drive train. The fuel cell reaction is moderated in a proton exchange membrane, which separates protons and electrons and rejoins them in the formation of water. Fuel-cell power has clear advantages over the diesel- and electric-power currently in use underground. Fuel-cell powered vehicles do not emit fumes like their diesel counterparts, nor must they lug around heavy lead-acid batteries or trail long power cords like electric mine vehicles. The Sandia designed and manufactured propulsion source stores its hydrogen as a hydride and meters out hydrogen as needed.

Laser-Induced Vaporization of Soot Investigated Using Time-Resolved Elastic Scattering

Laser-induced incandescence (LII) has become a widely used method to measure the amount of soot in combustion systems, ranging from fundamental burners to diesel engines. However, for the detected LII signal to be proportional to the volume fraction of soot, a number of conditions must be satisfied. One of these conditions is that the soot particle volume loss by vaporization be negligible. Because it is common practice in LII imaging experiments to use a laser fluence comfortably above the level needed for nascent vaporization (to assure that all particles in the field-of-view reach the vaporization temperature), reliable measurements require that particle volume loss by vaporization is either insignificant or known. Using a technique he calls time-resolved LIVES (laser-induced vaporization with elastic scattering), Pete Witze has shown that particle volume loss from LII can be significant.

LIVES is based on the idea that changes in soot volume fraction from laser-induced vaporization can be computed from the ratio of time-resolved measurements of elastic scattering from the soot and the incident laser pulse. (Details of the technique can be found in P. O. Witze, S. Hochgreb, D. Kayes, H. A. Michelsen, and C. R. Shaddix, Applied Optics, 40, 2443-2452 (2001)). The LIVES experimental setup uses a pulsed Nd:YAG laser to vaporize the soot and a fast transient recorder to capture the response of two photodiodes: one that monitors the incident laser pulse shape and one that measures elastically scattered light from the soot probe volume.

An example of LIVES measurements and analysis is shown in Figure 1 for a relatively high laser fluence of 0.72 J/cm^2 (nascent vaporization occurs at about 0.2 J/cm^2). The results reveal a large loss in soot volume fraction from laser-induced vaporization.

The abscissa used for the results presented in Figure 1a is time. However, it is more useful to plot the data as a function of the partial fluence (the accumulated fluence at any time within the laser pulse) as shown in Figure 1b for a range of total laser fluences. These results show that volume loss from laser-induced vaporization strongly depends on the rate of energy deposition, and not simply the partial fluence.

Pete believes that measurements of this type can be used to investigate the volatile organic fraction (VOF) contribution to diesel and gasoline particulate emissions. He is currently working on developing the technique for application to diesel exhaust in a project funded by the DOE Office of Transportation Technologies.

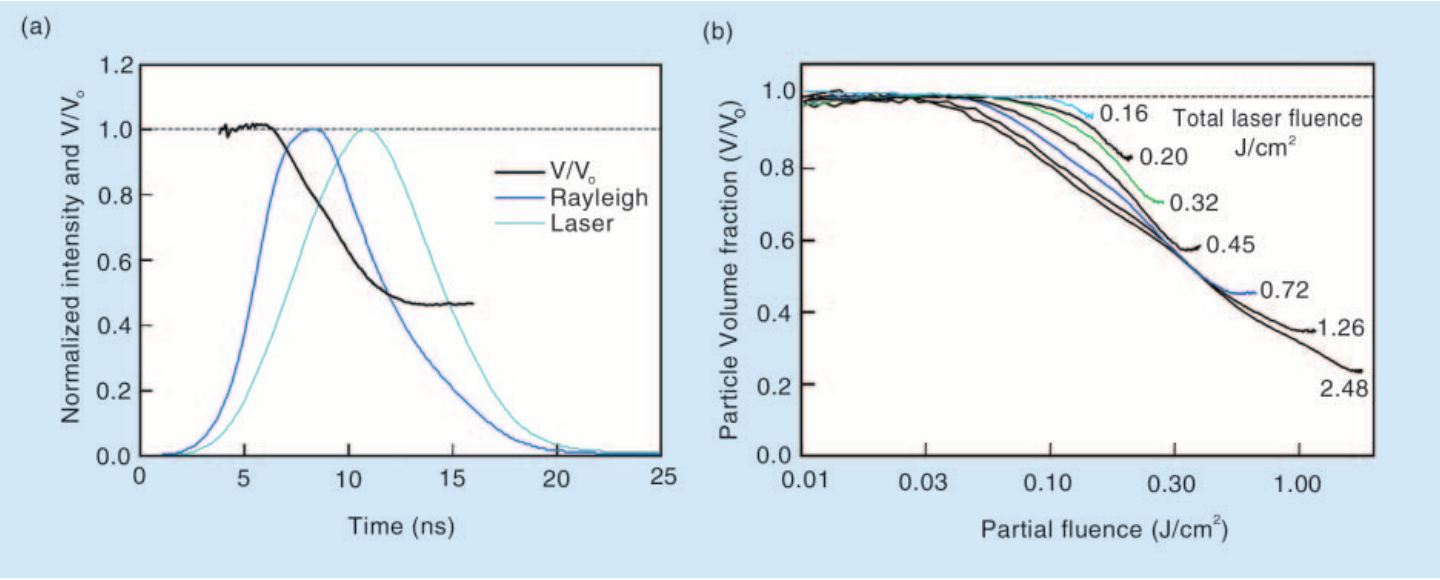

Figure 1. (a) Time-resolved measurements of laser intensity, elastic scattering, and particle volume fractions (V/V$_o$) inferred from the elastic scattering signal for a laser fluence of 0.72 J/cm^2. The success of these measurements depends on precise phasing of the scattering signal with the record of the incident laser pulse. (b) Particle volume fraction (V/V$_o$) reduction as a function of the partial laser fluence (log$_{10}$ scale) for different values of total laser fluence (total pulse). The data show that soot particle ablation depends both on the partial and total laser fluence and, by inference, the irradiance or rate of energy deposition.

Model Examines Catalytic Extension of Extinction Limits for Experimental Microcombustor

Small and stable combustors are currently being investigated because they could potentially find use as energy sources in microsystems, on-chip chemical detectors such as flame ionization devices, and microreactors and micropropulsion devices. For example, since the energy density of common fuels is 50 or more times that of the best high-output batteries, a tiny fuel tank coupled to a microcombustor could potentially replace several bulky batteries. Motivated by these considerations, Sandians Steve Margolis (SNL/CA) and Tim Gardner (SNL/NM) have developed a simple model of a prototype microcombustor.

Because they typically have large overall activation energies, premixed flames generally can only exist within certain parameter ranges, or extinction limits. These limits correspond to a rate of heat production that is sufficient to sustain the combustion reaction. In very small reactors, heat losses diminish the extinction limit because of the relatively large surface-to-volume ratio. However, catalysts can be used to extend these limits by lowering the activation energy. The prototype device built by Tim Gardner (SNL/NM), shown in Figure 1, utilizes a surface catalyst to enhance combustion and extend the extinction limit. Experimentally, it was found that combustion could be sustained in small volumes on the order of a couple of cubic millimeters.

An idealized geometry based on the conditions of the experiment and corresponding to a premixed flame in stagnation-point flow was used to model the effects of catalysis in extending the extinction limits of nonadiabatic stretched flames (Figure 2).

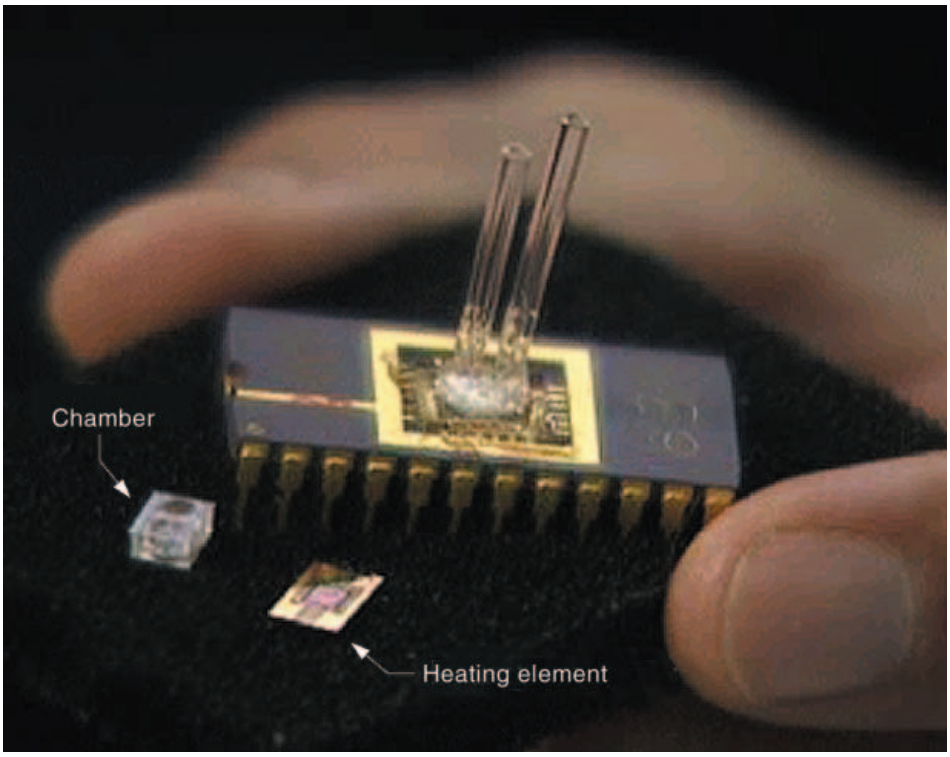

Figure 1. The prototype microcombustor was used to test the feasibility of using combustion to power microsystems. The glass tubes were for fuel entry and exhaust. The actual combustion chamber and heating element are shown next to the chip. This configuration was modeled to see if a catalyst could extend the life of the flame.

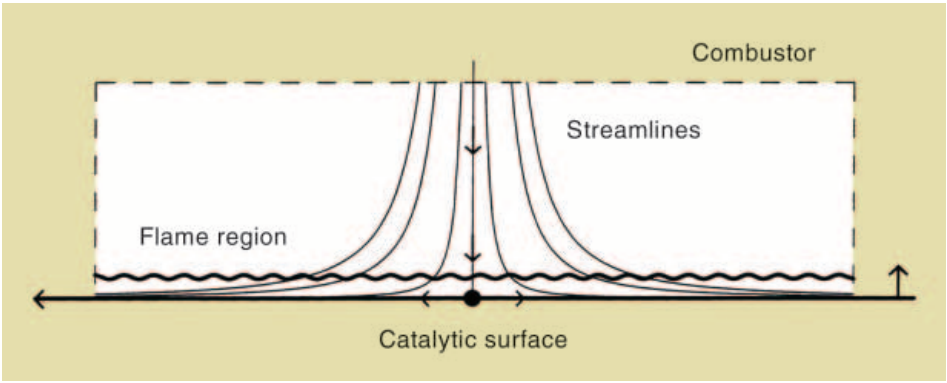

Figure 2. A schematic of the idealized catalyst-assisted stagnation-point geometry of the microcombustor. Heat loss was modeled volumetrically and is related to the surface-to-volume ratio of the combustor. Near extinction, the reaction region lies adjacent to the catalytic surface.

Specifically, a surface catalytic reaction was assumed to occur on the stagnation plane, thereby augmenting combustion in the bulk gas with an exothermic surface reaction characterized by a reduced activation energy.

Based on the fact that the activation energies remain large, an asymptotic analysis of the resulting flame structure yielded a formula for the extinction limit as a function of various parameters. In particular, the typical results illustrated in Figure 3 show that for parameter values that reflect those of the experiment, the presence of a surface catalyst extends the burning regime, thus counterbalancing the effects of heat loss and flame stretch that tend to shrink it. The analysis is relevant to small-volume combustors in general, where the increased surface-to-volume ratio can lead to extinction of the nonadiabatic flame in the absence of a catalyst.

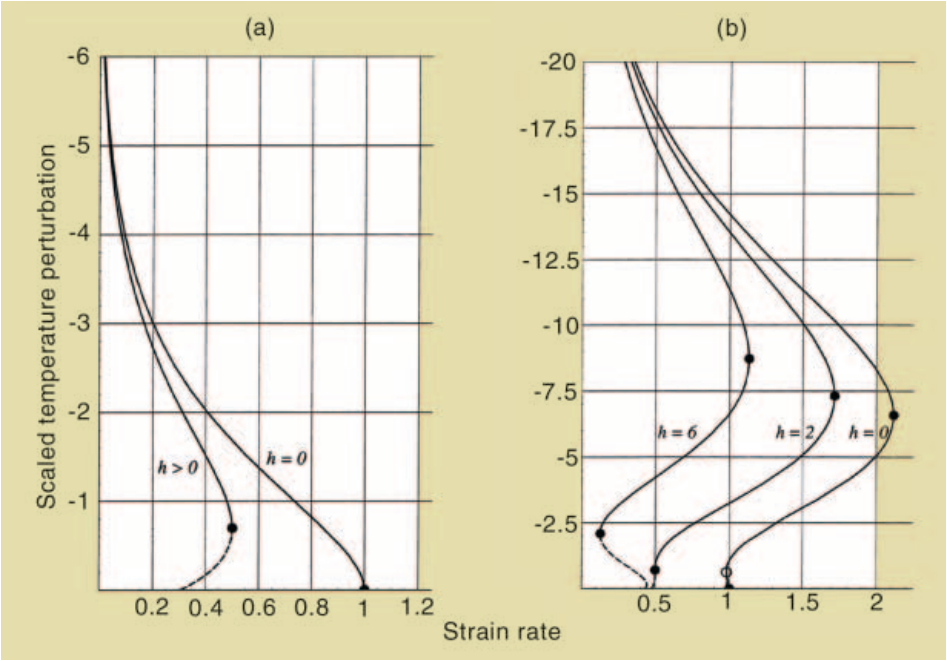

Figure 3. Solutions of the asymptotic combustion model for various conditions show how the presence of catalyst in the combustor could extend the extinction limit. In absence of catalyst (a) the solution is monotonic, whereas in solution (b) with catalyst, it can become nonmonotonic, resulting in an extension of the extinction limits from the lower to the upper set of sold circles. The strain rate is a critical parameter associated with extinction, while h is the heat loss parameter. The vertical axis (scaled temperature perturbation) is a scaled derivation of the surface temperature relative to a reference flame temperature.

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